Increasing the Length Scale in Temperature Accelerated Dynamics Simulations

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Molecular dynamics (MD) simulation is a powerful tool for probing systems in full atomistic detail. A general limitation, however, has been that the time scale is limited to nanoseconds, while many of the processes we wish to study take place over much longer times. Examples include film growth, bulk diffusion, and radiation damage annealing. Over the last few years, we have been developing *accelerated molecular dynamics* methods [1]. These methods extend the accessible simulation times for infrequent-event systems such as those mentioned above. Built on statistical mechanics principles, they give faster dynamical evolution from state to state without any prior assumptions about the atomistic mechanisms. The most powerful of these methods is temperature accelerated dynamics (TAD) [2], in which an elevated temperature is employed to speed the rate of transitions at the desired, lower temperature. Although direct high temperature dynamics would lead to an incorrect evolution from state to state, in the TAD method, the incorrect transitions are prevented, so that only the transitions that should occur at the lower temperature are accepted. When activation barriers are high relative to the temperature, TAD can give very large computational boost factors. For example, simulating vapor deposition of a metal film (e.g., Cu/Cu(100) at T = 77K [1]), gives a computational boost in the millions, allowing contact with the experimental deposition rate of monolayers per minute. These simulations exhibit surprisingly complicated mechanisms involving concerted motion of many atoms.

A serious limitation of TAD, however, is that only relatively small systems (e.g., ~10^3 atoms) can be treated. The computational scaling with the number of atoms, N, is roughly $N^{1.5}$ - N^3, depending on the ratio of the temperatures and the details of the implementation. This prevents TAD from being applicable to many important systems that have a natural length scale of thousands of atoms or more. In an effort to solve this problem, we are developing a way to spatially parallelize TAD by building on the synchronous sublattice algorithm (SSA) recently proposed by Shim and Amar [3] for parallelizing kinetic Monte Carlo simulations. In SSA, each processor is responsible for a spatial patch of the system, and each patch is further divided into subpatches, as shown in Fig. 1. Dynamical evolution is performed on one subpatch at a time, on all processors simultaneously. Cycling through the subpatch lattices gives very accurate evolution of the system provided the time slice for each subpatch is shorter than or comparable to the shortest event time. In the TAD implementation, each processor performs dynamics on a region larger than the subpatch, but only events centered within the subpatch are allowed to be accepted. Provided the subpatches are large enough to allow complex concerted events, the accuracy of the TAD method is retained.

In our first tests, we are studying the scaling behavior for the simple surface diffusion system shown in Fig. 2, in which the basic cell displayed in aqua is replicated to create a system of a desired size. Figure 3 shows preliminary timing results for both serial (nonparallel) TAD and the new SSA-TAD. The SSA-TAD has more overhead, but dramatically improved scaling is evident, allowing simulation of much larger systems than we have been able to do previously.

Moreover, for systems such as this that are free of long-range elastic relaxation, this scaling may flatten completely at larger sizes, so the accessible length scale would be limited only by the number of available processors. This would represent a significant breakthrough.

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[2] M.R. Sorensen and A.F. Voter, *J. Chem. Phys.* **112**, 9599 (2000).
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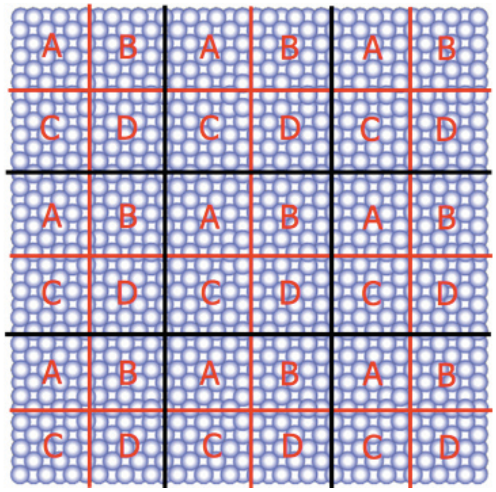

Fig. 1.
Schematic illustration of the synchronous sublattice algorithm (SSA) for spatially parallelizing kinetic Monte Carlo or temperature accelerated dynamics (TAD). A 9-processor case is shown.

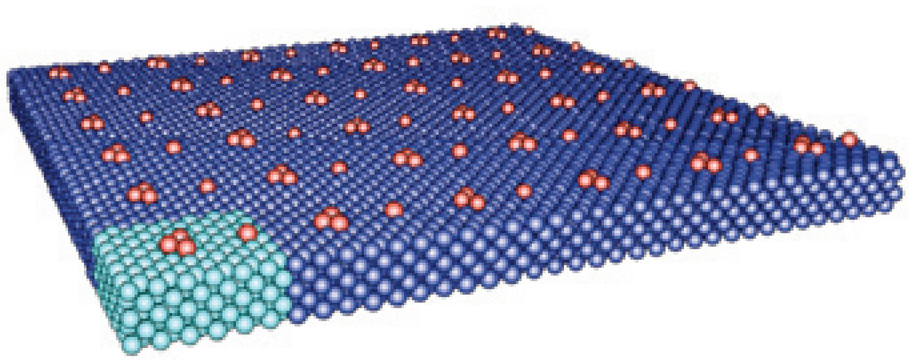

Fig. 2.
Cu/Cu(100) surface diffusion test system for SSA-TAD scaling tests. A 6x6 case for 36 processors is shown, while the unit cell (1x1) is indicated by aqua coloring.

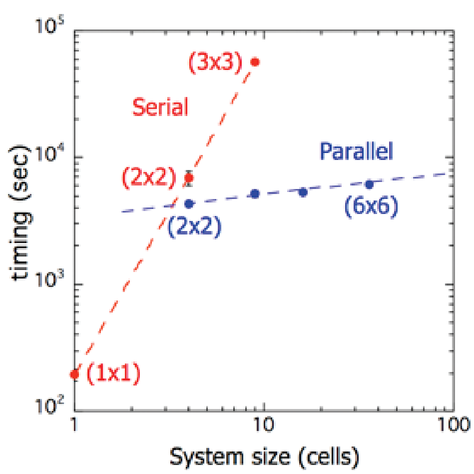

Fig. 3.
Timing results for TAD (red) and parallel SSA-TAD (blue) for propagation of the Cu/Cu(100) system at T = 100K for 0.01 s, using a high temperature of T = 600 K.